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PCT/CA2004/002111 Flora Tak Tak NG et al. Attorney Docket No. 04004.0160

## IAP20 Rec'd PCT/PTQ 09 JUN 2006

## AMENDMENTS UNDER PCT ARTICLE 34 (ARTICLE 34 AMENDMENTS)

International Application No.: PCT/CA2004/002111

MAIL STOP - PCT

Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450

Sir:

## REQUEST FOR SUBSTITUTION OF REPLACEMENT SHEETS

Please substitute the attached replacement sheets 54-61, containing the Article 34 Amendments to the claims filed February 22, 2006, for sheets 54-60, of the previously filed Article 34 Amendments to the claims filed June 6, 2005. It is respectfully requested that the claims contained in replacement sheets 54-61, be examined during examination of the patent application. Claims 1-47 are currently pending.

Respectfully submitted,

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Dated: June 9, 2006

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EFC/FPD/blc

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AMENDED SHEETS ATTACHED TO AMENDMENT UNDER ARTICLE 34 OF THE PCT, DATED FEBRUARY 22, 2006

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We claim:

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- 1. A catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus, the catalytic composite comprising:
- a) a support structure, made of a non zeolite inorganic oxide, having a void fraction ranging from 0.30 to 0.95 and a surface area of from  $40 \text{ m}^2/\text{g}$  to  $500 \text{ m}^2/\text{g}$ , the support structure having a shape selected from a ring, a hollow cylinder, a cross or multi partition ring or cylinder with 2, 3, or 4 cell partitions, a saddle, a solid ring, a solid cylinder, a sphere, and a honeycomb body; and
- b) from 0.01 to 10% by weight of a catalytically active species comprising a group VIII metal, based on the weight of the catalytic composite, which is deposited on the support structure.
  - 2. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to claim 1, wherein the void fraction is from 0.30 to 0.95 and the surface area is from  $50 \text{ m}^2/\text{g}$  to  $500 \text{ m}^2/\text{g}$ .
  - 3. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to claim 1 or 2, wherein the inorganic oxide is selected from the group consisting of alumina, silica, titania, zirconia and mixtures thereof.
  - 4. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to claim 1 or 2, wherein the inorganic oxide is

- 5. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to claim 1 or 2, wherein the inorganic oxide is  $\alpha$ -alumina.
- 6. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to any one of claims 1 to 5, wherein the support structure is in the shape of a Raschig ring.
- 7. The catalytic composite for use as a random packing
  material and catalyst in a catalytic distillation apparatus
  according to any one of claims 1 to 6, wherein the group
  VIII metal is nickel.
  - 8. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to any one of claims 1 to 7, wherein the group VIII metal is in the form of a metal salt or a metal complex.

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- The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus
   according to claim 8, wherein the metal salt is in an ionic state.
  - 10. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to claim 8, wherein the metal salt is a metal sulphate, a metal phosphate, a metal oxalate or a metal acetate.
  - 11. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to any one of claims 1 to 6, wherein the catalytically active species is nickel sulphate.

- 12. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to any one of claims 1 to 6, wherein the catalytically active species is nickel chloride.
- 5 13. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to any one of claims 9 to 12, wherein the catalytically active species is in admixture with ammonium sulphate or ammonium phosphate.
- 10 14. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to any one of claims 1 to 6, wherein the catalytically active species comprises a group VIII metal and a ligand, wherein the ligand comprises one or 15 more atoms selected from the group consisting of carbon, hydrogen, oxygen, nitrogen and phosphorus.
  - 15. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to claim 14, wherein the group VIII metal is in the zero oxidation state.

- 16. The catalytic composite for use as a random packing material and catalyst in a catalytic distillation apparatus according to any one of claims 1 to 6, wherein the group VIII metal is palladium, platinum or rhodium.
- 25 17. A process for the selective dimerization of a lower alkene to a  $C_6$ - $C_{12}$  alkene, which process comprises contacting the lower alkene with a catalytic composite as claimed in any one of claims 1 to 16, under catalytic distillation conditions.

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18. The process according to claim 17, wherein the lower alkene is selected from 1-butene, 2-butene and isobutene, and the  $C_6$ - $C_{12}$  alkene is selected from trimethylpentene, n-octene, dimethylhexene and methylheptene.

- 19. The process according to claim 17 or 18, wherein the catalytic composite is admixed with inert distillation packing.
- 20. The process according to claim 19, wherein the ratio of the catalytic composite to inert distillation packing is from 10:1 to 1:10.
  - 21. The process according to claim 19, wherein the catalytic composite and inert distillation packing are used in separate zones of the catalytic distillation column.
- 15 22. The process according to claim 17, wherein the lower alkene is a  $C_4$  alkene and the  $C_6$  to  $C_{12}$  alkene is predominantly a  $C_8$  alkene.
  - 23. The process according to claim 22, wherein the  $C_8$  alkene is a trimethylpentene.
- 24. A process for the hydrogenation of an alkene to an alkane, which process comprises contacting the alkene with a catalytic composite as claimed in any one of claims 14 to 16, and hydrogen, under catalytic distillation conditions.
- 25. The process according to claim 24 wherein the 25 alkene is selected from trimethylpentene, n-octene, dimethylhexene and methylheptene.
  - 26. The process according to claim 24 or 25, wherein the catalytic composite is admixed with inert distillation packing.

- 27. The process according to claim 26, wherein the ratio of the catalytic composite to inert distillation packing is from 10:1 to 1:10.
- 28. The process according to claim 26 wherein the catalytic composite and inert distillation packing are used in separate zones of the catalytic distillation column.

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- 29. The process according to any one of claims 24 to 28, wherein the alkene is trimethylpentene and the alkane is trimethylpentane.
- 30. A process for preparing high octane compounds, the process comprising:
  - a) contacting a  $C_2$  to  $C_6$  alkene with a catalytic composite as claimed in any one of claims 1 to 16, under catalytic distillation conditions, to obtain a  $C_6$  to  $C_{18}$  alkene; and
  - b) contacting the  $C_6$  to  $C_{18}$  alkene from step a) with a catalytic composite as claimed in any one of claims 14 to 16, and hydrogen, under catalytic distillation conditions, to obtain a  $C_6$  to  $C_{18}$  alkane.
- 20 31. The process according to claim 30, wherein the process steps a) and b) are carried out in a single catalytic distillation column.
  - 32. The process according to claim 30, wherein the process steps a) and b) are carried out in separate catalytic distillation columns.
  - 33. The process according to claim 30 or 31, wherein the  $C_2$  to  $C_6$  alkene is a  $C_4$  alkene and the  $C_6$  to  $C_{18}$  alkene is a  $C_8$  alkene.

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- 34. The process according to claim 33, wherein the  $C_8$  alkene is trimethylpentene.
- 35. A process for preparing high octane compounds, the process comprising:
- a) contacting isobutene with a catalytic composite as claimed in any one of claims 1 to 16, under catalytic distillation conditions, to obtain trimethylpentene; and
- b) contacting trimethylpentene with a
   hydrogenation catalyst, and hydrogen, under batch reaction
   conditions or under hydrogenation reaction conditions to
   obtain trimethylpentane.

- 36. A process for the production of  $C_6-C_{18}$  alkenes, which process comprises contacting a mixture of  $C_2-C_6$  alkenes with a catalytic composite as claimed in any one of claims 1 to 16, under catalytic distillation conditions.
- 37. A process according to claim 36, wherein each  $C_2$ - $C_6$  alkene in the mixture is oligomerized within different reactive zones found in a single catalytic distillation column.
- 20 38. A process according to claim 36, wherein each  $C_2$   $C_6$  alkene is oligomerized within different reactive zones found in two or more linked catalytic distillation column.
  - 39. A process according to any one of claims 36 to 38, wherein the mixture of  $C_2$ - $C_6$  alkenes comprises one or more  $C_4$  alkenes.
    - 40. A process for the selective oligomerization of a lower alkene to a  $C_6-C_{18}$  alkene, which process comprises contacting a mixture of  $C_2$  to  $C_6$  alkenes and  $C_1$  to  $C_6$

alkanes with a catalytic composite as claimed in any one of claims 1 to 16, under catalytic distillation conditions.

41. A catalytic composite for use as a random packing hydrogenation catalyst in a catalytic distillation apparatus, the catalytic composite comprising:

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- a) a support structure, made of an inorganic oxide and having a void fraction ranging from 0.30 to 0.95, the support structure having a shape selected from a ring, a hollow cylinder, a cross or multi partition ring or cylinder with 2, 3, or 4 cell partitions, a saddle, a solid ring, a solid cylinder, a sphere, and a honeycomb body; and
- b) from 0.01 to 10% by weight of palladium, platinum or rhodium, based on the weight of the catalytic composite, which is deposited on the support structure.
- 15 42. The catalytic composite for use as a random packing hydrogenation catalyst in a catalytic distillation apparatus according to claim 41, wherein the inorganic oxide is  $\alpha$ -alumina.
- 43. The catalytic composite for use as a random packing hydrogenation catalyst in a catalytic distillation apparatus according to claim 42, wherein the  $\alpha$ -alumina has a surface area of from 0.1 to 1.0 m<sup>2</sup>/g.
  - 44. A process for the hydrogenation of butadiene, the process comprising contacting butadiene with a catalytic composite as claimed in any one of claims 41 to 43, and hydrogen, under catalytic distillation conditions.
    - 45. A process for the selective hydrogenation of methylacetylene and propadiene in a C3 fraction to provide propylene, the process comprising contacting the C3

fraction with a catalytic composite as claimed in any one of claims 41 to 43, and hydrogen, under catalytic distillation conditions.

- 46. A process for the selective hydrogenation of allene and propyne in a fluid catalytic cracking (FCC) stream, the process comprising contacting the FCC stream with a catalytic composite as claimed in any one of claims 41 to 43, and hydrogen, under catalytic distillation conditions.
- 10 47. A process for the selective hydrogenation of butadiene in a raffinate I or a raffinate II stream to provide a butene, the process comprising contacting the raffinate I or the raffinate II stream with a catalytic composite as claimed in any one of claims 41 to 43, and hydrogen, under catalytic distillation conditions.